Multireference algebraic diagrammatic construction theory for accurate simulations of UV/Vis and X-ray absorption spectra of strongly correlated molecules

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Ab initio computational methods serve a critical role in the interpretation of many experimental photochemical processes, with the aim of understanding how the physical properties of molecules may change in response to absorbing light of a particular wavelength. Algebraic diagrammatic construction (ADC) theory is a routinely used class of many such theoretical methods, as it achieves a good balance of computational cost and accuracy in calculating the excited state properties of molecules. However, ADC theory has been shown to be unreliable when simulating excitation energies of molecules that are known to exhibit strong electron correlation in their ground and excited states. In this work, we present the strict and extended second-order formulations of multireference ADC (MR-ADC(2) and MR-ADC(2)-X), which combine the descriptions of static and dynamical electron correlation to accurately predict the excitation energies and excited state properties of strongly-correlated molecules. We describe the theoretical framework of these implementations and present an extensive benchmark of these methods for calculating the UV/Vis and X-ray absorption spectra of many weakly- and strongly correlated systems.